

# 1. PORTSMOUTH, OHIO RECYCLED URANIUM MASS BALANCE PROJECT

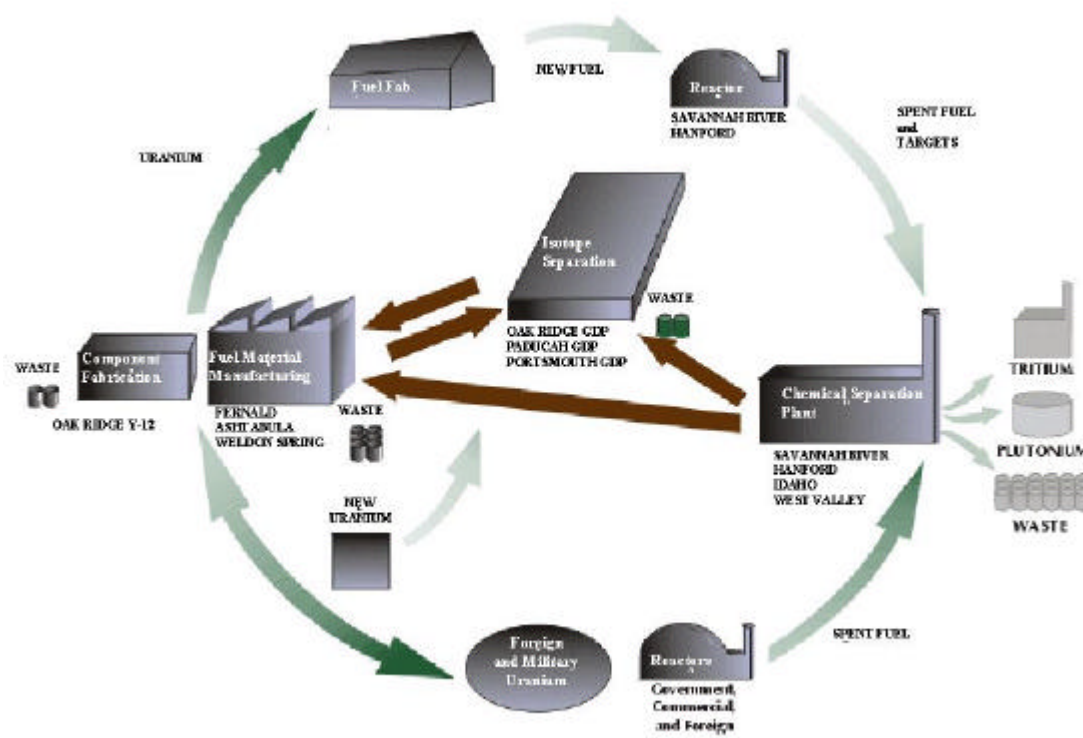
## 1.1 Project Overview

The gaseous diffusion plants (GDP) were an integral part of the flow path for uranium reprocessed from spent fuel from plutonium (Pu) and tritium production reactors. See Figure 1.1-1. Issues were raised surrounding this activity at the PGDP as to its potential for having affected the health of workers through exposure to the constituents of the RU. The Office of Environment, Safety and Health (EH) initiated five projects to investigate these legacy issues at the GDP's and linked plants. The second of these projects involves conducting a review of the characteristics and flow of uranium throughout the Department of Energy. This project is under the auspices of the Office of Nuclear Safety (EH-3) and is referred to as the mass balance project. It is the mass balance activity for PORTS that is addressed with this report.

The Bechtel Jacobs Company, LLC, under prime contract to DOE, was directed to prepare the PORTS site report for inclusion in the overall mass balance project report. A team consisting of seven long-term experienced current and former site and contractor employees with a cumulative service of 185 years was organized to review and research records of activities and RU operations. The team divided the site into four principal focus areas for directing its investigation. These areas represented the principle facilities and/or processes having the potential for concentrating the constituents of RU. Additionally, these facilities would present the greatest likelihood for worker interaction with uranium-bearing TRU/FP.

Figure 1.1-1

Principal Flow Streams of the Uranium Processing Cycle



These focus areas are: (1) uranium tetrafluoride ( $UF_4$ ) to  $UF_6$  feed manufacturing plant; (2) cascade and feed facilities; (3) uranium recovery; and (4) oxide conversion. Other facilities are addressed only to the extent that they were considered potential contributors to the mass balance issue. Activities at each of these facilities that involved either RU or the constituents of RU were reviewed from initial introduction of uranium during plant startup in FY 1955 through March 1999.

Four primary sources of RU were identified that bound the issue for Portsmouth. These sources were: (1) PGDP/ORGDP  $UF_6$  manufactured from usually depleted RU- $UO_3$ ; (2) PGDP product; (3) RU- $UF_6$  supplied from foreign sources; and (4) RU non- $UF_6$  supplied from many sources in small quantities. By tracking each of these four sources with time throughout each of the facilities and summing by facility, an annual inventory of RU constituents was created.

Data sources were researched to determine quantity, source, and transaction date of all uranium, regardless of form received or shipped at PORTS. A database of this information was prepared as the initial phase of this effort. Attempts to corroborate shipments and receipts with other sites were made for the principal RU shippers and receivers.

Classification of flows as RU was accomplished based on the following criteria:

1. Analytical data supports the presence of TRU or FP
2. Materials were of the characteristic enrichment levels of RU
3. Suppliers identified the materials as RU
4. Materials were coded as RU in Source and Special (SS) Accountability Reports

For this report, RU was assumed to maintain its identity as RU until it was fed. From that point on, the constituents of the RU were tracked. This approach was adopted since small amounts of RU were intermixed with much larger amounts of non-RU yielding product and tails streams containing de minimus quantities of TRU constituents.

Processing of RU was found to have occurred in three of the four primary facilities studied. No record could be found of any RU based materials ( $UF_4$ ) having ever been processed during the 46-1/2 months of X-344 feed manufacturing operation.

In the cascade, 1,094.6 MTU of RU was introduced as feed made at PGDP/ORGDP from depleted  $UO_3$ . Other foreign and domestic sources supplied 1.8 MTU of RU- $UF_6$ . Some of this material was fed as late as FY 1998. The PORTS Oxide Conversion Facility manufactured 1.9 MTU of RU- $UF_6$  (manufactured by various feed sources) that was fed to the cascade. The cascade concentrated the TRU constituent, neptunium (Np) and small quantities of Pu, at or near the RU feed points.

An estimated total of 60 to 90 kg of the fission product  $^{99}Tc$  was fed into the cascade, over the life of the plant, from low concentrations in large quantities of Paducah Product Feed (PPF). The cascade concentrates  $^{99}Tc$  in the top purge area and in top vent stream traps. A trap that was installed to remove  $^{99}Tc$  from the cascades' area of peak  $^{99}Tc$  concentration, concentrated  $^{99}Tc$  in the trap media.

In uranium recovery, 38.2 MTU was recovered in the form of triuranium octoxide  $U_3O_8$  from all sources during the period covered by this report. This oxide contained TRU (primarily Np) that had been removed from the cascade equipment during the two major cascade equipment change-out programs. The  $^{99}Tc$  concentrates in the sludges produced during the uranium recovery process.

In oxide conversion, 233 MTU of  $UF_6$  was produced, over approximately 20 years of operation, from various on-site and off-site (including foreign) sources. Of this production, 5.6 MTU was identified as having been made from RU source materials. The process of oxide conversion is shown to concentrate TRU primarily in filter ash, and to a lesser degree, in tower ash and trap media.

Adding all of the quantities of TRU/FP constituents over time bounds this issue at PORTS. Less than 0.3 g of Pu were estimated as received at the plant. Approximately 0.003 g entered the process equipment. A total of approximately 140g of Np were received with about 46 g entering the process equipment. Some 60 to 90 kg of <sup>99</sup>Tc must be considered as having been processed with much of this having been removed through venting to the environment or through sludges originating from uranium recovery operations or with the product stream. The quantities of TRU/FP constituents that did not enter the process equipment remained in the cylinders.

Worker exposure analysis consisted of a review of available workplace monitoring data that included TRU results and reports which summarized workers' exposure monitoring results of the In-Vivo and urine bioassay programs.

The greatest potential for worker exposure was determined by reviewing the operations where concentrations of TRU/FP occurred.

In the cascade, worker exposures may have occurred as:

1. Maintenance evolutions, including change-out of equipment at the RU feed points; and
2. Maintenance evolutions, including changes in the top purge and <sup>99</sup>Tc collection traps.

For uranium recovery, worker exposures may have occurred in the handling of the sludges and perhaps UF<sub>6</sub> cylinder washing operations. For oxide conversion, worker exposures may have occurred with the handling of the filter and tower ashes.

Since plant start up, many cases of worker exposure to and uptake of uranium are known and documented to have occurred. While no internal dose has been assigned to workers from TRU constituents of RU, it is likely that an uptake of these constituents has occurred at very low levels in the range of the limits of detection. Workers are known to have been exposed to <sup>99</sup>Tc.

## **1.2 Purpose and Scope**

The purpose of this project is to quantitatively estimate the historical mass flows and characteristics of RU within the PORTS site and between PORTS and other sites. The information generated from this project will enable the DOE to assess the potential for worker exposure and environmental contamination at PORTS resulting from the RU streams, specifically that caused by the TRU isotopes of Pu and Np and the fission product <sup>99</sup>Tc. These constituents were known to be present in trace amounts in uranium that had been recycled from DOE reactor programs and other sources. This project focuses on:

1. Identifying the mass flow of DOE RU from startup to March 31, 1999, from receipt to ultimate disposition. An intersite flow sheet was created showing how the PORTS site interfaced with other sites in the flow of RU. The flow includes all types of uranium including depleted, normal and enriched in several forms. The chemical forms specific to the site include uranium oxides (UO<sub>3</sub>, U<sub>3</sub>O<sub>8</sub>) and uranium dioxide (UO<sub>2</sub>), uranium fluorides (UF<sub>4</sub> and UF<sub>6</sub>), and uranium wastes that contain amounts of uranium that would affect mass balance studies;
2. Identifying the major facilities where the various forms of RU were received, processed, or treated, thereby concentrating the various TRU and FP constituents. The processes and activities are sufficiently described, including feed and product specifications, and the uranium streams characterized as to their content of TRU and FP to permit addressing worker or public health and safety issues; and
3. Performing a site mass balance to the degree existing mass and analytical data permit.

Items specifically excluded from this study and the rationale for exclusion is as follows:

1. Radioactive sources and standards.

These items are usually in sealed configurations or are in laboratory reagents. Their isotopic masses are accounted for under either the nuclear materials control and accountability system or the source control system. Their use is and has been controlled to assure worker safety and, as such, are not considered relevant to this study.

2. Very Highly Enriched (VHE) UF<sub>6</sub> shipments.

In order to assure that this effort remains unclassified, materials in this flow were not considered. Due to the behavior of TRU/FP in the gaseous diffusion cascade, it is likely that only <sup>99</sup>Tc may have been a constituent in this stream. Due to the extremely high alpha levels of the VHE uranium, the fractional contribution of any <sup>99</sup>Tc to dose calculations would be minimal.

3. Uranium Management Center inventories.

Materials were received after March 1999 and, therefore, are out of the project scope. These materials, however, are merely being stored with minimal worker interaction.

### 1.3 Project Implementation Strategy

The project goals are to:

1. Identify the mass flow of DOE RU from early production to March 31, 1999, including ultimate use or disposition;
2. Identify the characteristics and contaminants in the major uranium streams, specifically Pu, Np, and <sup>99</sup>Tc or other isotopic constituents of concern to worker or public health and safety (includes waste and scrap streams); and
3. Conduct PORTS site specific mass balances sufficiently thorough to identify significant implications for potential employee exposure to environmental contamination.

The strategy for accomplishing the PORTS mass balance project includes:

1. Utilizing existing DOE, Bechtel Jacobs Company, LLC, and United States Enrichment Corporation (USEC) protocols, procedures, and controls;
2. Obtaining and utilizing necessary “staff” specialists and support personnel through contractual means;
3. Establishing a structured approach to meeting the project goals, including the use of key assumptions;
4. Ensuring effective communication of progress, issues, and problem resolution through regular meetings with project personnel ; and
5. Coordinating with other sites and sharing of results.

This strategy is implemented through an organizational structure and a hierarchy of work elements described in Sections 1.3.1 and 1.3.2, respectively.

### 1.3.1 Project Organization

The mass balance project is planned and implemented through a matrix chain of responsibility and authority as shown in Figure 1.3.1-1.

The Office of Nuclear Safety within the DOE has overall responsibility for conducting this project as part of its plan to review the characteristics and flow of uranium throughout the DOE complex. The DOE headquarters team provides overall project direction and compiles the complex-wide report with assistance from the working group team leaders. Working group teams consisting primarily of DOE headquarters federal staff, are designated for each site to validate site data and results and assist in resolving any discrepancies between sites on shipper/receiver data, as well as provide assistance, as necessary, to complete the final site report. A data analysis subteam assists the DOE headquarters team in the analysis and consolidation of site report data for the final complex-wide report.

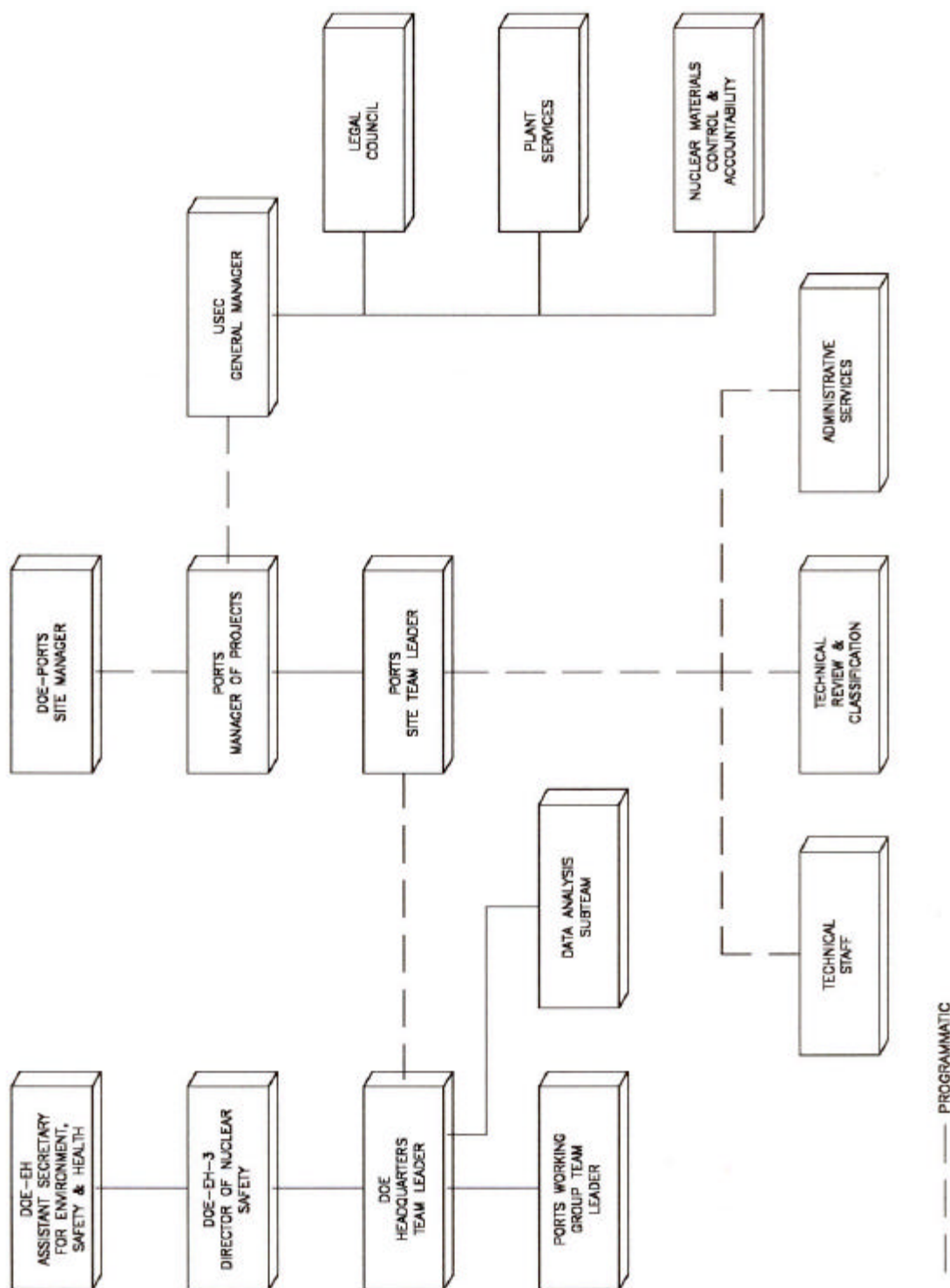
The PORTS site team is lead by a senior Bechtel Jacobs Company, LLC manager reporting to the PORTS Manager of Projects. The site team is composed of a subcontractor-based technical staff possessing over 185 years of combined experience in the maintenance, operations, engineering, analytical laboratory, and health and safety areas of the Portsmouth site. Support services, covering primarily nuclear materials control and accountability and records management, are provided, as needed, by the USEC. The site team is responsible for obtaining and summarizing site-specific RU data over the site's history, including mass flow and balance, TRU/FP constituent data, site inventory as of March 31, 1999, and for identifying major facilities/processes contributing to potential worker exposure.

### 1.3.2 Project Work Plan

Appendix B of the DOE Project Plan (Ref. 1) provides the foundation for the site's review of historical documents, extraction and evaluation of relevant data and preparation of this report. The framework to accomplish this project is shown in Figure 1.3.2-1. Data collection and evaluation were based upon the following key project assumptions:

1. Materials are classified as RU if:
  - a. Analytical data showed a positive indication for TRU;
  - b. Identified as such by the shipper;
  - c. Supplied as UF<sub>4</sub> or UF<sub>6</sub> at the characteristic RU enrichment (~0.63-0.68% <sup>235</sup>U);
  - d. Coded as RU in the source and special accountability records.
2. Materials that contained the fission product <sup>99</sup>Tc without significant accompanying quantities of TRU are not considered as RU (i.e., Paducah Product Feed);
3. Once RU materials enter into the cascade, or other process they are considered to have lost their RU identity;
4. Quantification of TRU constituents in UF<sub>6</sub> cylinders from PGDP and ORGDP and their subsequent feed rates are assumed to be in accordance with the Historical Impact of Reactor Tails on the Paducah Cascade (Ref 2, and 2a);
5. TRU/FP behavioral assumptions in the PORTS diffusion cascade are as follows:
  - a. The minute amount (if any) of Pu that enters the cascade lodges in the immediate vicinity of the feed point;

# PORTS MASS BALANCE PROJECT ORGANIZATIONAL STRUCTURE



# PORTS MASS BALANCE PROJECT WORK BREAKDOWN STRUCTURE

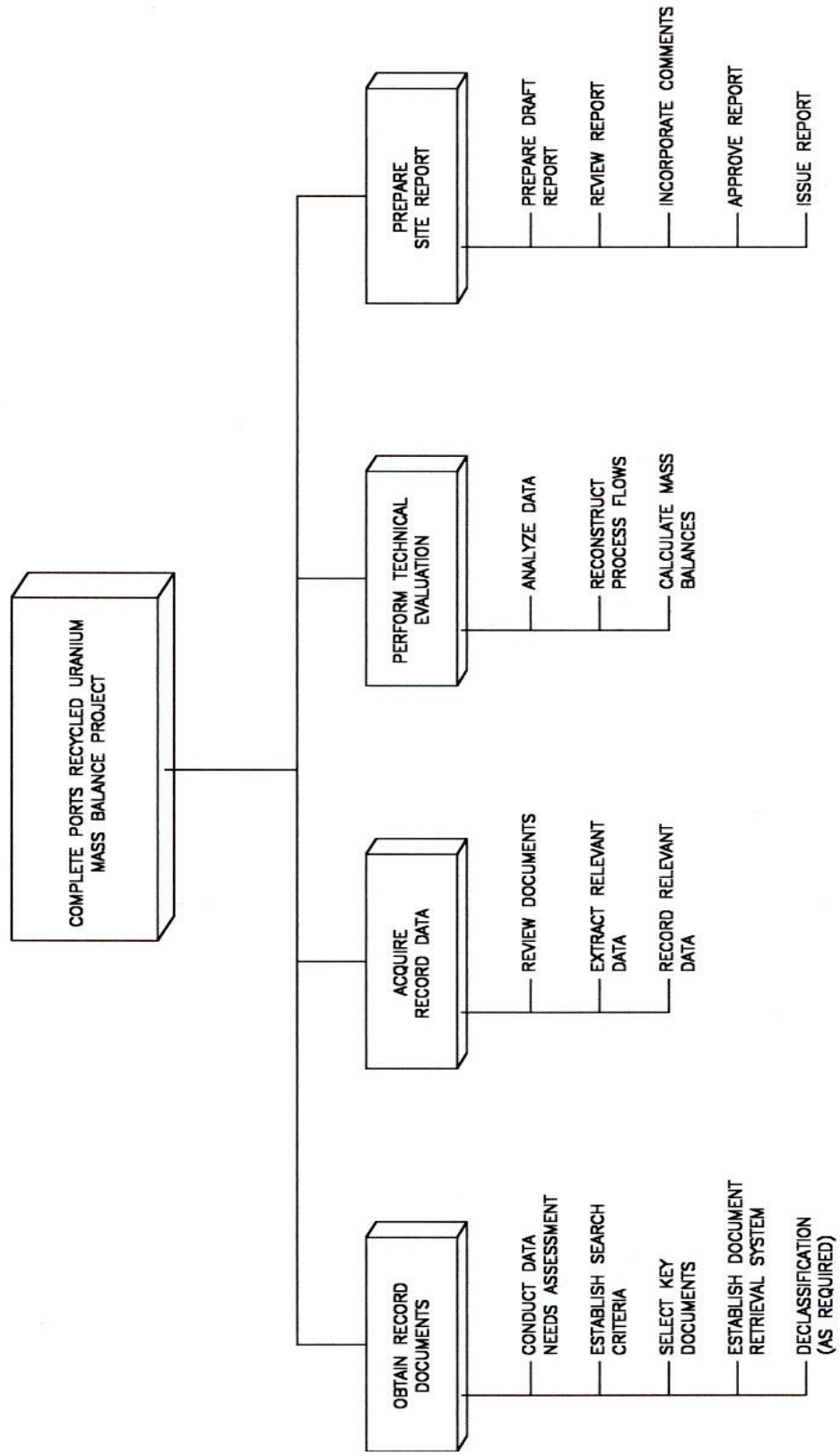


Figure 1.3.2-1

- b. Np entering the cascade becomes lodged on barrier and unplated surfaces close to the feed point until its removal during an equipment change-out; and
  - c. <sup>99</sup>Tc, due to its strong affinity for metallic surfaces, starts plating out at the feed point and then progresses up stream. The <sup>99</sup>Tc continues sorbing on metallic surfaces in successively higher cascade cells until the entire cascade above the feed point reaches equilibrium. The <sup>99</sup>Tc will then migrate to the top of the plant due to its lighter molecular weight.
6. A process is considered to have the ability to concentrate TRU/FP if it increases the mass of TRU/FP constituents relative to uranium or decreases the uranium mass relative to the amount of TRU/FP.